Wavelength Dependence of Aerosol Light Absorption

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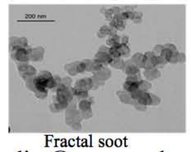

Fractal soot

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Abstract

There are some underappreciated theoretical results, and overlooked experimental results, for the Ångström coefficient for aerosol light (Å) absorption used to parameterize the wavelength dependence of light absorption. Å values are typically calculated using measurements or theory at two or more wavelengths, one typically in the near IR, and one in the near UV. Å is needed for remote sensing applications and to model aerosol evolution and climate impacts. The simplest model considers fractal-like soot particles composed of monomers that are much small than the wavelength of light. These fluffy fractal soot particles, considered with constant refractive index variation with wavelength, are thought to have an Å = 1 because absorption is the sum of that due to each tiny monomer. However, investigators such as Bond, Clarke, Kirchstetter, Lewis, Gyawali, Huebert, Hand, and coworkers report measurements of Å in the range of 0 to 5 for various wavelength pairs. Small values (i.e., Å < 1) of Å are often attributed to experimental error, or to the nebulous effects of large particles. Values of Å considerably larger than unity are often explained by considering the effects of light absorbing organic carbon coatings on particles. Our own measurements of Å using photoacoustic measurements of light absorption at 405 nm and 870 nm exhibit values of Å in the range from 0 to 5 for various combinations of wood smoke in the laboratory and in the ambient, as well as routine measurements dissected to scrutinize the diurnal variation. Mid afternoon is often associated with the smallest values of Å in urban settings relatively free of biomass burning, while the morning rush hour often produces values of Å near unity. What is going on? Sure enough, organic coatings that preferentially absorb light at shorter wavelengths on elemental carbon cores will, in general, be associated with values of Å significantly larger than unity as has frequently been observed, especially for biomass burning aerosol. However, it is underappreciated that a coating that doesn't absorb light at all, which may cause the fractal chain structure of the elemental carbon core to collapse to a more compact spherical shape, can induce values of Å as large 1.6, and as small as 0, depending on the size of the elemental carbon core. In other words, it is not only light absorbing organic carbon that causes values of Å larger than unity, but it is also due to the relatively enhanced light absorption at shorter wavelengths compared to longer wavelengths by coatings that might not significantly absorb light at all. A sufficiently large elemental carbon core certainly will cause values of Å near zero, and even negative values for monodispersions. Our poster summarizes our theoretical work with the coated sphere code, and measurements from campaigns in many cities and laboratories. We present our work in the context of others, and seek to clarify and address the observed variation of Å. We also find that filter-based measurements of Å using the Aethalometer may underestimate its true value due to wavelength dependent aspects of light absorption by particles on filters.

Sun Photometer and Photoacoustic

Sun photometry is very convenient for the measurement of optical depth of the atmosphere, Fig (1). The Langley plot method is applied in the process of calculation. The photoacoustic spectrometer is a non filter based instrument where the absorption is measured using the photoacoustic method and scattering is measured using reciprocal nephelometry, Fig (2) and (3)

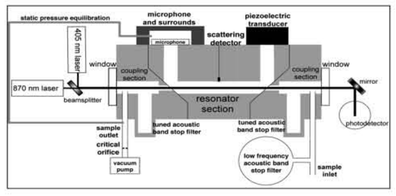

Fig (1). LED based six channel sun photometer operating at 430 nm, 470 nm, 530 nm, 660 nm, 870 nm, 950 nm.

Fig (2). Schematic of the dual wavelength photoacoustic instrument (Lewis, et al.,2008) operating at 405 nm and 870 nm for the measurement of aerosol light absorption and scattering.

Aethalometer

Aethelometer is a filter based instrument which measures optical absorption in real time and converts optical absorption to mass concentration of black carbon.

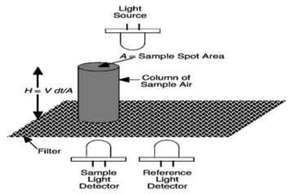

Fig (4). Schematic of light absorption measurement by means of filter in Aethelometer (Arnott, et al, 2005)

Modeling

Coated sphere theory was used to investigate the theoretically possible variations on Å for uncoated and coated carbon spheres. In this model the spherical carbon cores are supposed to be uniformly coated by organic and inorganic materials (*Toon and Ackerman*, 1981).

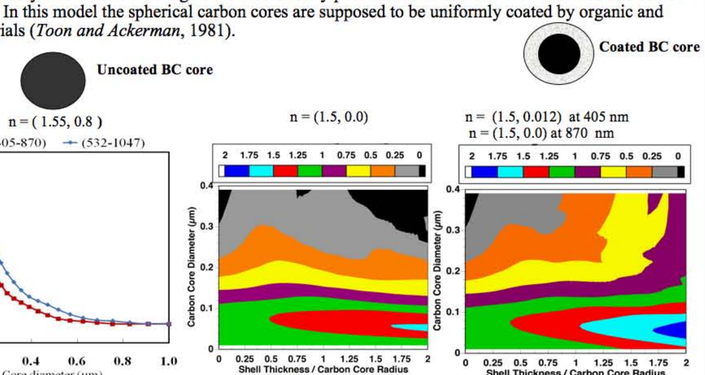

Fig 5. Ångström exponent of absorption for monodisperse uncoated carbon core of different diameter

Fig 6. Ångström exponent of absorption (405-870) for non absorbing coating(left) and absorbing coating (right) carbon core of different diameter.

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Results: Aerosol light absorption and scattering measurements at 405 nm and 870 nm in July and August in 2008 at Reno, NV

Fig 7. NASA satellite image of California wildfires of summer 2008. The satellite map shows the smoke extending from northern California to Reno, Nevada on July 10, 2008.(Courtesy of Jeff Reid)

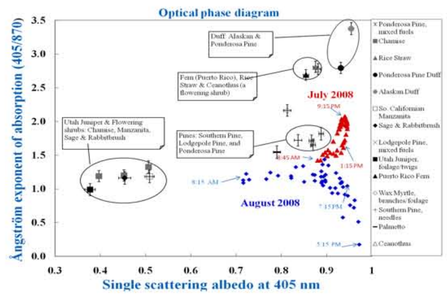

Fig 8. A comparison of monthly and diurnally averaged Å vs single scattering albedo (SSA) at 405 nm for the months of July and August 2008 with the similar optical properties of laboratory burned fuels. It illustrates that the optical properties of California wildfires aerosol of 2008 are similar to those of pine burning aerosol.

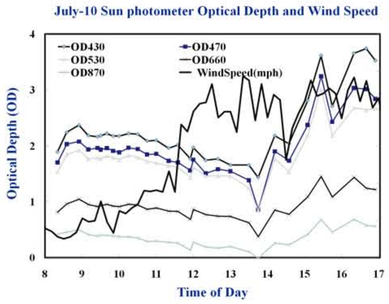

Fig 9. The spectral optical depth was very large for July-10, 2008. Also note the anti correlation between the wind speed and OD.

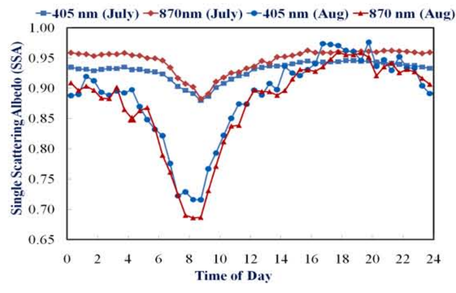

Fig 10. The monthly and diurnally averaged aerosol SSA for 405 nm and 870 nm for the months of July and August 2008. SSA is higher for the smoky month of July. Note the minimum around the "morning rush hour" due to the higher vehicular emission. The light absorbing organic carbon (in 2008) causes the SSA at 405 nm to be lower than 870 nm in July.

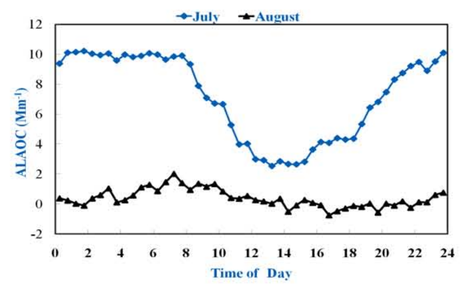

Fig 11. The monthly and diurnally averaged apparently light absorbing organic carbon (ALAOC) of July and August 2008. Note ALAOC is maximum for July and none for August. ALAOC Absorption = 405nm Abs - 870nm Abs (870/405).

Results: Measurements by Photoacoustic (PA) and Aethelometer (AE) in 2003 Jan-Feb at Las Vegas , NV

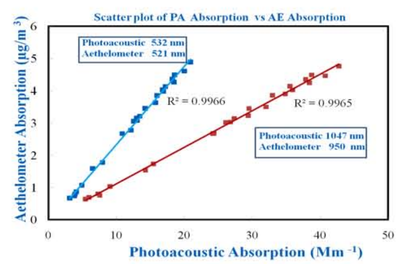

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Fig 12. The scatter plot of PA and AE measurements show very close agreement. Each point is part of the two months long diurnal average.

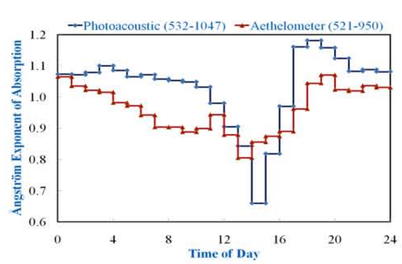

Fig 13. The comparison of Å measurement by PA and AE. AE absorption measurements are altered by filter matrix artifacts especially when organic coatings are present. The Å variation is less than that of in August for Reno.

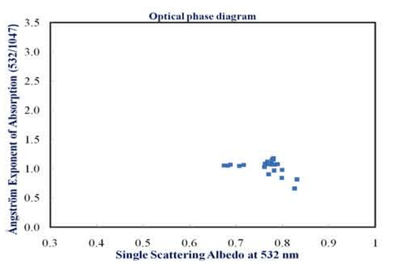

Fig 14. Å versus SSA for PA measurements. Note that Å significantly less than unity was also observed in Las Vegas as in Reno, NV. Compare with Fig 8.

Related Work

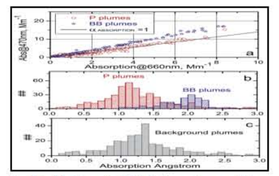

Fig 15. (a) Plot of PSAP absorption at 470 nm vs. 660 nm for BB (Biomass) and P (Pollution) revealing the difference in slope, (Clarke et al, 2007). Histograms of Å for BB and for P plumes (b) and for non plume (c). Note specially the large Å for BB and , Å < 1 for P.

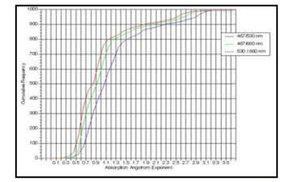

Fig 16. Frequency of occurrence of aerosol Å, by PSAP in GoMACCS field campaign. Note 80% data has Å around one, and 20% has Å from 1.5 to 3. Some of the data have Å less than 1, (R. W. Bergstrom et al. 2007)

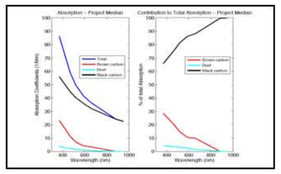

Fig 17. Apportioning of total light absorption to black carbon, brown carbon and dust for different wavelengths, (M. Yang, 2008).

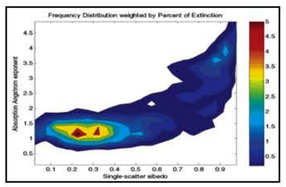

Fig 18. Frequency plot of the optical properties of emissions from traditional wood burning cook stove, (A. R. Christoph et al, 2008).

Note : In all these related works we are interested in the observations of the varying possible values of Å especially less than one in different situations and the role of Biomass, ALAOC, Black carbon, dust and mixing state of aerosols for absorption of radiation.

Conclusions

- The simulations confirm that large Å values are possible even when coatings do not absorb light. The organic coating need not be intrinsically brown to observe effects referred to those caused by brown carbon.
- In the optical phase diagram the Å values are significantly less than one in Reno and Las Vegas during the later part of the day, and are nearly one for the rush hour.
- Although the scatter plot of PA and AE measurements show closer agreement, the comparison of the Å shows that PA measurements vary more at some parts of the day especially when organic and inorganic coating of black carbon dominates.
- Wildfires are a major source of the apparently light absorbing organic carbon at 405 nm.
- The optical properties of the summer 2008 California wildfires aerosol closely resemble that of the pine smoke aerosols from lab measurements. Å vary from 1.3 to 2.3, an indication of highly wavelength dependent absorption.
- The monthly average of hourly variation of SSA significantly decreases from 7 AM to 12 AM (for the rush hour) for July and August, indicating the strong presence of vehicular emission of black carbon and organic carbon.